

MAILED 05 JAN 1995

FORM PTO-1390 (REV 5-93)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			Mo-4188/LeA 29,111
INTERNATIONAL APPLICATION NO. PCT/EP 93/01651			U.S. APPLICATION NO. (if any) 08/362547
INTERNATIONAL FILING DATE June 28, 1993		PRIORITY DATE CLAIMED July 9, 1992	
TITLE OF INVENTION A PROCESS FOR THE PRODUCTION OF RIGID FOAMS CONTAINING URETHANE GROUPS AND PREDOMINANTLY ISOCYANURATE GROUPS			
APPLICANT(S) FOR DO/EO/US 1) Rolf Wiedermann, 2) Stephan Wendel, 3) Wolfgang Schmitz			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Items 11. to 16. below concern other document(s) or information included:</p> <p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. Form PTO 1449 and References</p> <p>12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input type="checkbox"/> Other items or information:</p>			

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Donna J. Veatch

(Name of person mailing paper or fee)

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PATENT APPLICATION
Mo-4188
LeA 29,111 PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF)
ROLF WIEDERMANN ET AL)
SERIAL NUMBER: TO BE ASSIGNED)
FILED: HERewith)
TITLE: A PROCESS FOR THE PRO-)
DUCTION OF RIGID FOAMS)
CONTAINING URETHANE)
GROUPS AND PREDOMINATELY)
ISOCYANURATE GROUPS)

PCT/EP93/0165

2 1/2 A
D. Gassaway
4-20-95

PRELIMINARY AMENDMENT

The Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

Upon the granting of a Serial Number and filing date and prior to the
examination of the subject application, kindly amend the claims as follows:

IN THE CLAIMS:

- a' 1. (Amended) A process for the production of rigid foams containing
urethane groups and [predominantly] predominately isocyanurate groups by
reaction of
- 1) polyisocyanates with
 - 2) compounds containing at least two isocyanate-reactive hydrogen atoms and
having a molecular weight of 400 to 10,000 in the presence of

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- 3) organic blowing agents and
4) flameproofing agents and
5) compounds containing at least two isocyanate-reactive hydrogen atoms and having a molecular weight of 32 to 399 as crosslinking agent and optionally in the presence of
6) auxiliaries and additives known per se,
- characterized in that component 2) has branched chains and in that component 2), the flameproofing agent 4) and the crosslinking component 5) are used as a formulation in which component 2) is present in a quantity of 30 to 90 parts by weight, component 4) is present in a quantity of 10 to 60 parts by weight and component 5) is present in a quantity of 10 to 20 parts by weight, the parts by weight of these components adding up to 100, and in that the reaction is carried out in an index range of 200 to 600.

2. (Amended) [A] The process [as claimed in] of claim 1, [characterized in that] wherein C₁₋₆ hydrocarbons are used as the organic blowing agent.

3. (Amended) [A] The process [as claimed in claims 1 and] of claim 2, [characterized in that] wherein pentane is used as the organic blowing agent.

4. (Amended) [A] The process [as claimed in claims 1 and 2] of claim 1, [characterized in that] wherein cyclopentane is used as the organic blowing agent.

5. (Amended) [A] The process [as claimed in claims 1 to 4] of claim 1, [characterized in that] wherein a mixture of 15 to 50% by weight n- and/or isopentane and 85 to 50% by weight cyclopentane is used as the organic blowing agent.

6. (Amended) [A] The process [as claimed in claims 1 and 2] of claim 2, [characterized in that] wherein hexane is used as the organic blowing agent.

7. (Amended) [A] The process [as claimed in claims 1 and 2] of claim 1, [characterized in that] wherein cyclohexane is used as the organic blowing agent.

8. (Amended) [A] The process [as claimed in claims 1 and 2] of claim 1, [characterized in that] wherein mixtures of pentane, cyclopentane, hexane and/or cyclohexane are used as the organic blowing agent.

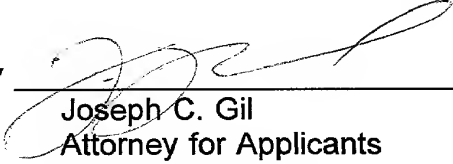
REMARKS

This amendment has been made to place the claims in better form according to U.S. practice.

Respectfully submitted,

ROLF WIEDERMANN
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By


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08/362547

A PROCESS FOR THE PRODUCTION OF RIGID FOAMS CONTAINING
URETHANE GROUPS AND PREDOMINANTLY ISOCYANURATE GROUPS

This invention relates to a process for the production of flameproofed, CFC-free rigid polyisocyanurate foams.

5 The production of halogen-free rigid polyisocyanurate foams using water and hydrocarbons as blowing agents is known. The use of water as blowing agent has the disadvantage that the formation of polyurea structures (in consequence of the reaction of isocyanate with water accompanied by the elimination of carbon dioxide) turns the surface of the foams brittle (surface brittleness) so that
10 adhesion between the foam and the surface skin is adversely affected.

Although, on the other hand, surface brittleness is reduced by using only hydrocarbons as blowing agent, the percentage content of flameproofing agents which normally
15 act as plasticizers has to be increased to ensure flame resistance, so that purely hydrocarbon-blown rigid foams are generally not dimensionally stable and show shrinkage/contraction.

It has now surprisingly been found that, without any
20 adverse effect on the required flame resistance, purely hydrocarbon-blown rigid polyisocyanurate foams do not shrink providing polyols having branched chains are used.

In the context of the invention, "branched" chains are understood to be linear chains from which one or more side
25 chains branch out; in addition to carbon atoms, heteroatoms may also be present in the linear chain and in the side chain. This definition also encompasses linear chains bearing methyl substituents.

The present invention relates to a process for the
30 production of rigid foams containing urethane groups and predominantly isocyanurate groups by reaction of

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Donna J. Veatch

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1) polyisocyanates with

2) compounds containing at least two isocyanate-reactive
hydrogen atoms and having a molecular weight of 400 to
10,000 in the presence of

3) organic blowing agents and

4) flameproofing agents and

5) compounds containing at least two isocyanate-reactive
hydrogen atoms and having a molecular weight of 32 to
399 as crosslinking agent and optionally in the
presence of

6) auxiliaries and additives known per se,

characterized in that component 2) has branched chains and
in that component 2), the flameproofing agent 4) and the
crosslinking component 5) are used as a formulation in
which

component 2) is present in a quantity of 30 to 90 parts by
weight,

component 4) is present in a quantity of 10 to 60 parts by
weight and

component 5) is present in a quantity of 10 to 20 parts by
weight,

the parts by weight of these components adding up to 100,
and in that the reaction is carried out in an index range
of 200 to 600.

In preferred embodiments of the invention,

- C₁₋₆ hydrocarbons are used as the organic blowing agent,
- pentane is used as the organic blowing agent,
- 5 - cyclopentane is used as the organic blowing agent,
- a mixture of 15 to 50% by weight n- and/or isopentane and 85 to 50% by weight cyclopentane is used as the organic blowing agent,
- 10 - hexane is used as the organic blowing agent,
- cyclohexane is used as the organic blowing agent,
- 15 - mixtures of pentane, cyclopentane, hexane and/or cyclohexane are used as the organic blowing agent.

The following starting components are used for the production of the rigid polyisocyanurate foams:

- 20
1. Aliphatic, cycloaliphatic, araliphatic, aromatic and heterocyclic polyisocyanates of the type described for example by W. Siefken in Justus Liebigs Annalen der Chemie, 562, pages 75 to 136, for example those
 - 25 corresponding to the following formula



in which

30 n = 2 - 4, preferably 2,
and

Q is an aliphatic hydrocarbon radical containing 2 to 18 and preferably 6 to 10 carbon atoms, a cycloaliphatic hydrocarbon radical containing 4

35 to 15 and preferably 5 to 10 carbon atoms, an

aromatic hydrocarbon radical containing 6 to 15 and preferably 6 to 13 carbon atoms or an arali-phatic hydrocarbon radical containing 8 to 15 and preferably 8 to 13 carbon atoms,

5

for example polyisocyanates of the type described in DE-OS 28 32 253, pages 10 - 11.

10 In general, particular preference is attributed to the polyisocyanates readily obtainable on an industrial scale, for example 2,4- and 2,6-tolyene diisocyanate and mixtures of these isomers ("TDI"); polyphenyl polymethylene polyisocyanates of the type obtained by phosgenation of aniline-formaldehyde condensates ("crude MDI") and carbodiimide-, urethane-,
15 allophanate-, isocyanurate-, urea- and biuret-modified polyisocyanates ("modified polyisocyanates"), more particularly modified polyisocyanates derived from 2,4- and/or 2,6-tolyene diisocyanate or from 4,4'-and/or 2,4'-diphenylmethane diisocyanate.

20

2. Other starting components are compounds with branched molecular chains which contain at least two isocya-
nate-reactive hydrogen atoms and which have a molecu-
lar weight of generally 400 to 10,000. In addition
25 to compounds containing amino groups, thiol groups or carboxyl groups, preferred compounds of this type are compounds containing hydroxyl groups, more especially 2 to 8 hydroxyl groups, particularly those having a molecular weight in the range from 1,000 to 8,000 and
30 preferably from 2,000 to 4,000, for example compounds containing at least two, generally two to eight and preferably two to four hydroxyl groups of the type known per se for the production of homogeneous and cellular polyurethanes, as described for example in
35 DE-OS 2 832 253, pages 11 to 18.

This starting component preferably contains up to 50% by weight polyester, based on the mixture as a whole.

- 5 3. The blowing agents used are organic blowing agents, preferably C₁₋₆ hydrocarbons, more preferably pentane, especially n- and/or isopentane, also cyclopentane and mixtures thereof with n- and/or isopentane.
- 10 4. The flameproofing agents used are flameproofing agents known per se, preferably products liquid at 20°C.
- 15 5. Other starting components are compounds containing at least two isocyanate-reactive hydrogen atoms and having a molecular weight in the range from 32 to 399. In this case, too, the compounds in question are compounds containing hydroxyl groups and/or amino groups and/or thiol groups and/or carboxyl groups, preferably compounds containing hydroxyl groups and/or amino groups which serve as crosslinking agents. These compounds generally contain 2 to 8 and preferably 2 to 4 isocyanate-reactive hydrogen atoms. Examples of such compounds can be found in DE-OS 28 32 253, pages 19 - 20.
- 20
- 25 6. Auxiliaries and additives known per se, such as emulsifiers and foam stabilizers, are optionally used. Preferred emulsifiers are those based on alkoxylated fatty acids and higher alcohols.
- 30 Suitable foam stabilizers are, above all, polyether siloxanes, particularly water-soluble types. These compounds generally have a structure in which a copolymer of ethylene oxide and propylene oxide is attached to a polydimethyl siloxane group. Foam stabilizers such as these are described, for example,
- 35

in US-PSS 2,834,748, 2,917,480 and 3,629,308. The catalysts known per se from polyurethane chemistry, such as tert. amines and/or organometallic compounds, may also be used.

5 Reaction retarders known per se, for example compounds showing an acidic reaction, such as hydrochloric acid or organic acid halides; cell regulators known per se, such as paraffins or fatty alcohols or
10 dimethyl polysiloxanes; pigments or dyes; stabilizers against ageing and weathering; plasticizers; fungistatic and bacteriostatic agents; and fillers, such as barium sulfate, kieselguhr, carbon black or whiting, may also be used.

15 Further examples of surface-active additives and foam stabilizers, cell regulators, reaction retarders, stabilizers, flameproofing agents, plasticizers, dyes, fillers, fungistatic and bacteriostatic agents which may optionally be used in accordance with the invention and information on the use of these additives and
20 the way in which they work can be found in Kunststoff-Handbuch, Vol. VII, edited by Vieweg and Höchtlen, Carl-Hanser-Verlag, München 1966, for example on pages 103 to 113.

25 The process according to the invention is carried out as follows:

 According to the invention, the reaction components are reacted by the single-stage process known per se, the prepolymer process or the semiprepolymer process, often
30 using machines, for example of the type described in US-PS 2,764,565. Particulars of processing machines which may also be used in accordance with the invention can be found in Kunststoff-Handbuch, Vol. VIII, edited by Vieweg and Höchtlen, Carl-Hanser-Verlag, München 1966, for example on
35 pages 121 to 205.

According to the invention, the reaction is carried out in the index range from 200 to 600 and preferably in the index range from 250 to 450.

5 According to the invention, foaming may also be carried out in closed molds. To this end, the reaction mixture is introduced into a mold. Suitable mold materials are metals, for example aluminium, or plastics, for example epoxy resin.

10 The foamable reaction mixture foams in the mold and forms the molding. The in-mold foaming reaction may be carried out in such a way that the molding has a cellular structure at its surface. However, it may also be carried out in such a way that the molding has a compact skin and a cellular core. According to the invention, it is possible in this regard to introduce a foamable reaction mixture into the mold in such a quantity that the foam formed just fills the mold. However, it is also possible to introduce more foamable reaction mixture into the mold than is necessary for filling the interior of the mold with foam. This technique is known as overcharging and is described, for example, in US-PSS 3,178,490 and 3,182,104.

15 "External release agents" known per se, such as silicone oils, are often used for in-mold foaming. However, so-called "internal release agents", as known for example from DE-OSS 2 121 670 and 2 307 589, may also be used, optionally in admixture with external release agents.

20 However, foams may also be produced by slabstock foaming or by the laminator process known per se.

30 The rigid foams obtainable in accordance with the invention are used for applications requiring high flame resistance, for example in the building industry, for insulation of the engine compartment of trucks and automobiles, as coating materials with high flame resistance and for the surface insulation of engine hoods as protection against noise.

35

Examples

Starting products

Polyol A (Comparison):

5 A mixture (formulation) of the following components was prepared:

- 100 parts by weight of a polyol mixture, OH value 198, consisting of
- 10 45 parts by weight of a polyether, OH value 185, prepared by reaction of ethylene glycol with ethylene oxide
- 8 parts by weight diethylene glycol, OH value 1060
- 5 parts by weight of a polyester prepared by reaction of phthalic anhydride with benzyl alcohol and butanol
- 15 27 parts by weight of the commercially available flame-proofing agent Disflamoll® DPK (a product of Ciba Geigy Plastics and Additives Co.)
- 15 parts by weight of a polyester, OH value 200, prepared by reaction of adipic acid and phthalic anhydride with diethylene glycol.
- 20

Polyol B (Invention):

 A mixture (formulation) of the following components was prepared:

- 25
- 100 parts by weight of a polyol mixture, OH value 168, consisting of:
- 27 parts by weight of the commercially available flame-proofing agent Disflamoll® DPK (a product of Ciba Geigy Plastics and Additives Co.)
- 30 12 parts by weight of a polyether, OH value 880, prepared by reaction of trimethylol propane with propylene oxide
- 50.5 parts by weight of a polyether, OH value 45, prepared by reaction of trimethylol propane with propylene
- 35

oxide and ethylene oxide

10.5 parts by weight of a polyether, OH value 380, prepared by reaction of trimethylol propane with propylene oxide.

5

Polyol C (Invention):

A mixture (formulation) of the following components was prepared:

- 10 100 parts by weight of a polyol mixture, OH value 212, consisting of:
- 27 parts by weight of the commercially available flame-proofing agent Disflamoll® DPK (a product of Ciba Geigy Plastics and Additives Co.)
- 15 13 parts by weight of a polyether, OH value 880, prepared by reaction of trimethylol propane with propylene oxide
- 40 parts by weight of a polyether, OH value 56, prepared by reaction of propylene glycol with propylene oxide
- 20 10 parts by weight of a polyether, OH value 380, prepared by reaction of trimethylol propane with propylene oxide
- 10 parts by weight of a polyester, OH value 370, prepared by reaction of adipic acid, phthalic anhydride, oleic acid and trimethylol propane.
- 25

Polyol D (Invention):

A mixture (formulation) of the following components was prepared:

30

- 100 parts by weight of a polyol mixture, OH value 215, consisting of:
- 27 parts by weight of the commercially available flame-proofing agent DEEP (diethyl ethyl phosphonate)
- 35 13 parts by weight of a polyester, OH value 370, prepared

- by reaction of adipic acid, phthalic anhydride, oleic acid and trimethylol propane
- 10 parts by weight of a polyether, OH value 880, prepared by reaction of trimethylol propane with propylene oxide
- 5 25 parts by weight of a polyester, OH value 210, prepared by reaction of adipic anhydride, phthalic anhydride, glycerol and propylene glycol
- 10 20 parts by weight of a polyether, OH value 56, prepared by reaction of propylene glycol with propylene oxide
- 25 parts by weight of a polyester, OH value 300, prepared by reaction of phthalic anhydride with diethylene glycol and ethylene oxide.

Table 1 (Comparison)

Formulation in parts by weight

Comparison Examples	1	2
Polyol A	98	98
Dimethyl cyclohexyl amine	0.14	0.18
Potassium acetate solution (1)	0.54	0.7
Cyclopentane	8	12.5
Stabilizer B1605 (Bayer AG)	2	2
Polyisocyanate (crude MDI, Desmodur® 44V20, a product of Bayer AG)	103	166
Index	219	351
Density [kg/m ³]	42	39
Adhesion of the paper covering layer after 24 hours	Good	Good
Ø Flame height in small burner test DIN 4102 [mm]	130	120
DIN 4102 classification	B2	B2
Dimensional stability	Shrinkage	(Less) Shrinkage
Surface brittleness	None	None

(1) 25% solution in diethylene glycol

The results in Table 1 show that, although surface brittleness can be completely eliminated by using only hydrocarbons as blowing agent in the foaming of polyols having unbranched molecular chains with polyisocyanates to form polyisocyanurate foams, the foams obtained are not dimensionally stable.

Table 2 (Invention)

Formulation in parts by weight

Examples

	1	2	3	4	5 (Comparison)
Polyol D	-	-	-	98	-
Polyol C	-	-	98	-	98
Polyol B	98	98	-	-	-
Dimethyl cyclohexyl amine	0.36	0.45	0.36	0.36	0.36
Potassium acetate solution (1)	1.4	1.75	1.4	1.4	1.2
Stabilizer B1605 (Bayer AG)	2	2	2	2	2
Cyclopentane	8	12.5	8.5	8.0	6.5
Polyisocyanate (crude MDI, Desmodur® 44V20 (Bayer AG))	103	166	116	105	70
Index	244	392	223	200	142
Density [kg/m ³]	42	39	42	42	42
Adhesion of the paper covering layer after 24 hours	Good	Good	Good	Good	Good
Ø Flame height in the small burner test acc. to DIN 4102 [mm]	135	135	130	135	140
DIN 4102 classification	B2	B2	B2	B2	B2
Dimensional stability	No shrinkage	No shrinkage	No shrinkage	Almost no shrinkage	No shrinkage
Surface brittleness	None	None	None	None	None

(1) 25% solution in diethylene glycol

The results of Examples 1 to 4 in Table 2 show that the polyols according to the invention achieve a DIN 4102 fire classification of B2 and are dimensionally stable where cyclopentane is used as the blowing agent. Example 5 in the Table shows that indices above 200 have to be applied.

CLAIMS

1. A process for the production of rigid foams containing urethane groups and predominantly isocyanurate groups by reaction of

1) polyisocyanates with

2) compounds containing at least two isocyanate-reactive hydrogen atoms and having a molecular weight of 400 to 10,000 in the presence of

3) organic blowing agents and

4) flameproofing agents and

5) compounds containing at least two isocyanate-reactive hydrogen atoms and having a molecular weight of 32 to 399 as crosslinking agent and optionally in the presence of

6) auxiliaries and additives known per se,

characterized in that component 2) has branched chains and in that component 2), the flameproofing agent 4) and the crosslinking component 5) are used as a formulation in which

component 2) is present in a quantity of 30 to 90 parts by weight,

component 4) is present in a quantity of 10 to 60 parts by weight and

component 5) is present in a quantity of 10 to 20 parts by weight,

Sub
a's
cont

the parts by weight of these components adding up to 100, and in that the reaction is carried out in an index range of 200 to 600.

2. A process as claimed in claim 1, characterized in that C₁₋₆ hydrocarbons are used as the organic blowing agent.

3. A process as claimed in claims 1 and 2, characterized in that pentane is used as the organic blowing agent.

4. A process as claimed in claims 1 and 2, characterized in that cyclopentane is used as the organic blowing agent.

5. A process as claimed in claims 1 to 4, characterized in that a mixture of 15 to 50% by weight n- and/or isopentane and 85 to 50% by weight cyclopentane is used as the organic blowing agent.

6. A process as claimed in claims 1 and 2, characterized in that hexane is used as the organic blowing agent.

7. A process as claimed in claims 1 and 2, characterized in that cyclohexane is used as the organic blowing agent.

8. A process as claimed in claims 1 and 2, characterized in that mixtures of pentane, cyclopentane, hexane and/or cyclohexane are used as the organic blowing agent.

Add B1

COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

A process for the production of rigid foams containing
urethane groups and predominantly isocyanurate groups

the specification of which (check one)

☐ is attached hereto

☒ was filed on June 28, 1993 as Application
Serial No. PCT/EP93/0165 and was amended on
_____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

PRIOR FOREIGN APPLICATION(S)

			Priority claimed	
			<input checked="" type="checkbox"/>	<input type="checkbox"/>
			Yes	No
<u>P 42 22 519.1</u>	<u>Germany</u>	<u>09.07.1992</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	(Day/month/year filed)		
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	(Day/month/year filed)	Yes	No
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	(Day/month/year filed)	Yes	No

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

(Application Serial No.) (Filing date) (STATUS: patented/pending/abandoned)

(Application Serial No.) (Filing date) (STATUS: patented/pending/abandoned)

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

JOSEPH C. GIL, Patent Office Registration Number 26,602
ARON PREIS, Patent Office Registration Number 29,426
LYNDANNE M. WHALEN, Patent Office Registration Number 29,457
THOMAS W. ROY, Patent Office Registration Number 29,582
RICHARD E. L. HENDERSON, Patent Office Registration Number 31,619
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N. DENISE BRWON, Patent Office Registration Number 36,097

all of Miles Inc., Pittsburgh, Pennsylvania 15205-9741.

SEND CORRESPONDENCE TO: <u>Patent Department</u> <u>Miles Inc.</u> <u>Mobay Road</u> <u>Pittsburgh, Pennsylvania 15205-9741</u>	DIRECT TELEPHONE CALLS TO: (Name and telephone number) <u>Patent Department</u> <u>(412) 777-2349</u>
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

FULL NAME OF SOLE OR FIRST INVENTOR <u>Rolf Wiedermann</u>	INVENTOR'S SIGNATURE <u>Rolf Wiedermann</u>	DATE <u>3 Nov. 1994</u>
RESIDENCE <u>51519 Odenthal-Voismwinkel, Germany</u>	<u>DEX</u>	CITIZENSHIP <u>German</u>
POST OFFICE ADDRESS <u>c/o Bayer Aktiengesellschaft, 51368 Leverkusen, Germany</u>		
FULL NAME OF SECOND JOINT INVENTOR, IF ANY <u>Stephan Wendel</u>	INVENTOR'S SIGNATURE <u>Stephan Wendel</u>	DATE <u>4 Nov. 1994</u>
RESIDENCE <u>51373 Leverkusen, Germany</u>	<u>DEX</u>	CITIZENSHIP <u>German</u>
POST OFFICE ADDRESS <u>c/o Bayer Aktiengesellschaft, 51368 Leverkusen, Germany</u>		
FULL NAME OF THIRD JOINT INVENTOR, IF ANY <u>Wolfgang Schmitz</u>	INVENTOR'S SIGNATURE <u>Wolfgang Schmitz</u>	DATE <u>8 Nov. 1994</u>
RESIDENCE <u>51519 Odenthal-Blecher, Germany</u>	<u>DEX</u>	CITIZENSHIP <u>German</u>
POST OFFICE ADDRESS <u>c/o Bayer Aktiengesellschaft, 51368 Leverkusen, Germany</u>		
FULL NAME OF FOURTH JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP
POST OFFICE ADDRESS		
FULL NAME OF FIFTH JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP
POST OFFICE ADDRESS		